

The following listing of the claims replaces all prior listings and versions of the claims in this application.

Listing of the Claims

1. (Previously Presented) A continuous process for preparing highly esterified polyol fatty-acid polyester by interesterifying polyol containing more than about four esterifiable hydroxy groups and fatty-acid ester of easily removable alcohol in a heterogeneous reaction mixture wherein
 - a) a catalyst is used in the reaction mixture at an initial level of from about 0.01 to about 0.5 mole of catalyst per mole of polyol;
 - b) a soap emulsifier is used in the initial stage of the process at a level of from about 0.001 to about 0.6 mole of soap per mole of polyol;
 - c) the molar ratio of total ester reactant to each esterifiable hydroxy group of the polyol in the reaction mixture ranges from about 0.9:1 to about 1.2:1;
 - d) the temperature in the initial stage of the process ranges from about 130°C to about 140°C, and in the final stages of the process ranges from about 80°C to about 120°C; and
 - e) easily removable alcohol is removed from the reaction mixture as the interesterifying reaction proceeds; and

wherein the initial stage of the interesterifying reaction is carried out in a continuous manner under conditions of backmixing suitable for maintaining within said reaction mixture a level of lower partial fatty acid esters of said polyol that is sufficient to emulsify said reaction mixture.

Claims 2-4 (Cancelled).

5. (Previously Presented) The process of Claim 1 in which the initial catalyst level is from about 0.01 to about 0.1 mole of catalyst per mole of polyol.

6. (Original) The process of Claim 5 in which the catalyst level is from about 0.02 to about 0.05 mole of catalyst per mole of polyol.

Claim 7 (Cancelled).

8. (Previously Presented) The process of Claim 1 wherein said initial level of soap emulsifier is from about 0.2 to about 0.4 mole per mole of polyol.

9. (Original) The process of Claim 8 wherein said soap emulsifier is a potassium soap of hydrogenated fatty acid containing from about 10 to about 22 carbon atoms.

Claims 10-12 (Cancelled).

13. (Previously Presented) The process of Claim 1 wherein said molar ratio of said total ester reactant to said esterifiable hydroxy group is from about 1:1 to about 1.2:1.

14. (Previously Presented) The process of Claim 62 wherein the initial stage of said interesterifying reaction is carried out under conditions of backmixing until the average degree of esterification of the polyol is from about 20% to about 70%, to thereby provide sufficient lower partial polyol polyester to aid in solubilization of the polyol.

15. (Previously Presented) The process of Claim 14 wherein the initial stage of said interesterifying reaction is carried out under conditions of backmixing until the average degree of esterification of the polyol is from about 35% to about 60%.

Claims 16-22 (Cancelled).

23. (Previously Presented) The process of Claim 1, wherein the temperature in said initial stage is between about 132°C and about 135°C.

Claims 24-26 (Cancelled).

27. (Previously Presented) The process of Claim 62 wherein, in the initial stage of the interesterifying reaction, the reaction mixture contains soap emulsifier at a level of from about 0.001 to about 0.6 mole per mole of polyol.

28. (Original) The process of Claim 27 wherein said soap emulsifier is at a level of from about 0.2 to about 0.4 mole per mole of polyol and said conditions of backmixing are continued until the degree of esterification of said polyol is from about 30% to about 60%.

Claim 29 (Cancelled).

30. (Previously Presented) The process of Claim 62 wherein the temperature in the initial stage is from about 132°C to about 135°C and the temperature in the subsequent stages is from about 100°C to about 120°C.

Claims 31-42 (Cancelled).

43. (Previously Presented) The process of Claim 62 wherein said molar ratio of said total ester reactant to each said esterifiable hydroxy group is from about 1:1 to about 1.2:1.

44. (Previously Presented) The process of Claim 62 wherein the final degree of esterification of said polyol reaches at least about 70%.

45. (Original) The process of Claim 44 wherein the final average degree of esterification of the polyol is at least about 95%.

Claims 46-47 (Cancelled).

48. (Previously Presented) The process of Claim 27 wherein the final stages of the reaction are carried out under plug-flow conditions, after the degree of esterification of said polyol has reached at least about 50%.

Claims 49-50 (Cancelled).

51. (Previously Presented) The process of Claim 48 wherein the molar ratio of said total ester reactant to each said esterifiable hydroxy group of said polyol is from about 1:1 to about 1.2:1.

Claims 52-53 (Cancelled).

54. (Previously Presented) The process of Claim 14 which is carried out in a series of at least two reaction vessels.

55. (Original) The process of Claim 54 wherein there are from three to about eight of said reaction vessels.

Claims 56-61 (Cancelled).

62. (Previously Presented) In a continuous process for preparing highly esterified polyol fatty acid polyester by interesterifying polyol containing more than four esterifiable hydroxy groups and fatty acid ester of an easily removable alcohol in a heterogeneous reaction mixture wherein said easily removable alcohol is removed from said reaction mixture as the reaction proceeds, the improvement which comprises:

- (A) carrying out an initial stage of the interesterifying reaction in a continuous manner under conditions of backmixing suitable for maintaining within said reaction mixture a level of lower partial fatty acid esters of said polyol that is sufficient to emulsify said reaction mixture; and

- (B) carrying out at least a final stage of the interesterifying reaction in a continuous manner under conditions approaching plug-flow conditions after the degree of esterification of said polyol has reached at least about 50%.

63. (Previously Presented) A process for the synthesis of polyol fatty-acid polyesters by reacting a polyol and a fatty-acid lower-alkyl ester under substantially solvent free conditions in the presence of a catalyst and an emulsifier, the process comprising an initial reaction stage which is carried out under such conditions that the reaction mixture in said initial stage is in steady-state, with continuous introduction of reactants comprising polyol and fatty-acid lower-alkyl ester, and continuous removal of products comprising reaction mixture having a degree of esterification of about 10% or more and volatile alcohol formed during the initial reaction stage, and one or more subsequent reaction stages in which the reaction mixture from said initial stage is further reacted to said polyol fatty-acid polyesters.

64. (Previously Presented) The process according to Claim 63 wherein the reaction mixture from the initial stage is further reacted to said polyol fatty acid polyesters after combining with any remaining part of the fatty-acid lower-alkyl ester reactant.

65. (Previously Presented) A process according to Claim 63 wherein the emulsifier is an alkali metal soap.

66. (Previously Presented) A process according to Claim 63 wherein the alkali metal soap is selected from the group of soaps having a chain length within the range of from 8 to 22 carbon atoms.

67. (Previously Presented) A process according to Claim 63 wherein the fatty-acid lower-alkyl ester is a fatty-acid methyl ester.

68. (Previously Presented) A process according to Claim 63 wherein the catalyst is selected from the group consisting of potassium hydroxide and carbonates of potassium and sodium.

69. (Previously Presented) A process according to Claim 63 wherein the reaction mixture in said initial reaction stage has a degree of esterification of within the range of from 10 to 60%.

70. (Previously Presented) A process according to Claim 63 wherein the reaction mixture in said initial stage does not contain any substantial amount of solvent.

71. (Previously Presented) A process according to Claim 63 wherein the reaction temperature in said initial stage is maintained at a level of within the range of from 130° to 140°C.

72. (Previously Presented) A process according to Claim 63 wherein the average residence time of the reaction mixture in said initial stage is caused to be about 1.5 hours.

73. (Previously Presented) A process according to Claim 63 wherein the molar ratio of catalyst to polyol in said initial reaction stage is within the range of from about 0.01:1 to about 0.5:1.

74. (Previously Presented) A process according to Claim 63 wherein the molar ratio of emulsifier to polyol in said initial reaction stage is within the range of from 0.2:1 to 0.6:1.

75. (Previously Presented) A process according to Claim 63 for the synthesis of polyol fatty-acid polyesters having a degree of esterification of at least about 70%.

76. (Previously Presented) A process according to Claim 63 wherein the polyol is sucrose.

77. (Previously Presented) A process according to Claim 76 wherein the molar ratio of fatty-acid lower-alkyl ester to sucrose is within the range of from 7.2:1 to 15:1.

78. (Previously Presented) A process according to Claim 63 wherein said initial reaction stage is fully separate from said one or more subsequent reaction stages.

79. (Previously Presented) A process according to claim 63, wherein the partial vapor pressure of the volatile alcohol in the initial reaction stage is less than 100 mm Hg.

80. (Previously Presented) A process according to claim 79, wherein the partial vapor pressure of the volatile alcohol is maintained by sparging with an inert gas.

81. (Previously Presented) A process according to claim 63, wherein the one or more subsequent reaction zones are provided in a tray reactor.

82. (Previously Presented) A process according to claim 63, wherein the emulsifier is used in the initial reaction stage in an amount of from about 3% to about 11% by weight of the reactants.

83. (Previously Presented) A process according to claim 63, wherein the initial reaction stage is carried out in a continuous reaction vessel having stirring means.

84. (Previously Presented) A process according to claim 83, wherein in the initial reaction stage the stirring means applies agitation to ensure thorough mixing of the reaction components.

85. (Previously Presented) A process according to claim 63, wherein the initial reaction stage is carried out in a continuous stirred tank reactor.

86. (Previously Presented) A process according to claim 63, wherein the steady-state reaction mixture in the first zone is capable of solubilizing the polyol.

87. (Previously Presented) A process according to claim 63, wherein the final degree of esterification is 95% or more.

88. (Previously Presented) A process according to claim 63, wherein the final degree of esterification is 98% or more.

89. (Previously Presented) A process for the synthesis of polyol fatty-acid polyesters by reacting a polyol and a fatty-acid lower-alkyl ester of a volatile C₁-C₄ alcohol under substantially solvent free conditions in the presence of a catalyst and an emulsifier, the process comprising:

an initial reaction stage which is carried out under such conditions that the reaction mixture in said initial stage is in steady-state, with continuous introduction of reactants comprising polyol and fatty-acid lower-alkyl ester, and continuous removal of products comprising reaction mixture having a degree of esterification of about 10% or more and volatile alcohol formed during the initial reaction stage, and

one or more subsequent reaction stages in which the reaction mixture from said initial stage is further reacted to said polyol fatty-acid polyesters.

90. (Previously Presented) A process according to claim 89, wherein the fatty-acid lower-alkyl ester is a fatty-acid methyl ester.

91. (Previously Presented) A process according to claim 89, wherein there is one in-going reactant stream in the initial stage.

92. (Previously Presented) A process according to claim 89, wherein the emulsifier is used in the initial reaction stage in an amount of from about 3% to about 11% by weight of the reactants.

93. (Previously Presented) A process for the synthesis of polyol fatty-acid polyesters by reacting a polyol and a fatty-acid lower-alkyl ester under substantially solvent free conditions in the presence of a catalyst selected from the group consisting of alkali metals, alloys of two or more alkali metals, alkali metal hydrides, alkali metal alkoxides, potassium carbonate, sodium carbonate, barium carbonate, potassium hydroxide and mixtures thereof, and an emulsifier, the process comprising:

an initial reaction stage which is carried out under such conditions that the reaction mixture in said initial stage is in steady-state, with continuous introduction of reactants comprising polyol and fatty-acid lower-alkyl ester, and continuous removal of products comprising reaction mixture having a degree of esterification of about 10% or more and volatile alcohol formed during the initial reaction stage, and

one or more subsequent reaction stages in which the reaction mixture from said initial stage is further reacted to said polyol fatty-acid polyesters.

94. (Previously Presented) A process according to claim 93, wherein the fatty-acid lower-alkyl ester is an ester of a volatile C₁-C₄ alcohol.

95. (Previously Presented) A process according to claim 93, wherein the catalyst is selected from the group consisting of sodium carbonate, potassium carbonate, potassium hydroxide and mixtures thereof.

96. (Previously Presented) A process according to claim 63, wherein the fatty-acid lower-alkyl ester is an ester of a volatile C₁-C₄ alcohol.

97. (Previously Presented) A process according to claim 96, wherein the catalyst is selected from the group consisting of alkali metals, alloys of two or more alkali metals, alkali metal hydrides, alkali metal alkoxides, potassium carbonate, sodium carbonate, barium carbonate, potassium hydroxide and mixtures thereof.

98. (Previously Presented) A process according to claim 97, wherein the molar ratio of fatty-acid lower-alkyl ester to esterifiable sites on the polyol is from about 0.9:1 to about 1.4:1.

99. (Previously Presented) A process according to claim 97, wherein soap is used in the initial reaction stage in an amount of from about 3% to about 11% by weight of the reactants.

100. (Previously Presented) A process according to claim 97, wherein the emulsifier introduced into the initial reaction stage is soap.

101. (Previously Presented) A process according to claim 100, wherein the polyol is sucrose and reaction mixture product from the initial stage comprises not more than 4.2% unreacted sucrose.

102. (Previously Presented) A process according to claim 101, wherein the initial reaction stage temperature is in the range of from about 130°C to about 140°C.

103. (Previously Presented) A process according to claim 102, wherein the initial reaction stage is carried out in a continuous stirred tank reactor.

104. (Previously Presented) A process according to claim 102, wherein the initial reaction stage is carried out in a continuous reaction vessel having stirring means.

105. (Previously Presented) A process according to claim 96, wherein the catalyst is selected from the group consisting of sodium carbonate, potassium carbonate, potassium hydroxide and mixtures thereof.

106. (Previously Presented) A process according to claim 96, wherein in the initial reaction stage the degree of esterification is between about 10% and about 70%.

107. (Previously Presented) A process according to claim 63, wherein the initial reaction stage is carried out in a continuous stirred reaction vessel having stirring means, wherein in the initial reaction stage the degree of esterification is between about 10% and about 70%, and wherein the fatty-acid lower-alkyl ester is an ester of a volatile C₁-C₄ alcohol.

108. (Previously Presented) A process according to claim 107, wherein in the initial reaction stage the stirring means applies agitation to ensure thorough mixing of the reaction components.

109. (Previously Presented) A process according to claim 108, wherein the initial reaction stage temperature is in the range of from about 130°C to about 140°C.

110. (Previously Presented) A process according to claim 85, wherein the fatty-acid lower-alkyl ester is an ester of volatile C₁-C₄ alcohol.

111. (Previously Presented) A process according to claim 110, wherein the catalyst is selected from the group consisting of alkali metals, alloys of two or more alkali metals, alkali metal hydrides, alkali metal alkoxides, potassium carbonate, sodium carbonate, barium carbonate, potassium hydroxide and mixtures thereof.

112. (Previously Presented) A process according to claim 111, wherein the initial reaction stage temperature is in the range of from about 130°C to about 140°C.

113. (Previously Presented) A process according to claim 112, wherein the emulsifier is soap.

114. (Previously Presented) A process for the synthesis of polyol fatty-acid polyesters by reacting a polyol and a fatty-acid lower-alkyl ester under substantially solvent free conditions in the presence of a catalyst selected from the group consisting of alkali metals, alloys of two or more alkali metals, alkali metal hydrides, alkali metal alkoxides, potassium carbonate, sodium carbonate, barium carbonate, potassium hydroxide and mixtures thereof, and an emulsifier, the process comprising:

an initial reaction stage wherein the temperature is in the range of from about 130°C to about 140°C, which is carried out under such conditions that the reaction mixture in said initial stage is in steady-state, with continuous introduction of reactants comprising polyol and fatty-acid lower-alkyl ester, and continuous removal of products comprising reaction mixture having a degree of esterification of about 10% or more and volatile alcohol formed during the initial reaction stage, and

one or more subsequent reaction stages in which the reaction mixture from said initial stage is further reacted to said polyol fatty-acid polyesters.

115. (Previously Presented) A process according to claim 114, wherein the emulsifier is used in the initial reaction stage in an amount of from about 3% to about 11% by weight of the reactants.

116. (Previously Presented) A process for the synthesis of polyol fatty-acid polyesters by reacting a polyol and a fatty-acid lower-alkyl ester under substantially solvent free conditions in the presence of a catalyst and an emulsifier, the process comprising:

an initial reaction stage which is carried out under such conditions that the reaction mixture in said initial stage is in steady-state, with continuous introduction of reactants comprising polyol and fatty-acid lower-alkyl ester, and continuous removal of products comprising reaction mixture having a degree of esterification of about 10% or more and volatile alcohol formed during the initial reaction stage, and

one or more subsequent reaction stages in which the reaction mixture from said initial stage is further reacted to said polyol fatty-acid polyesters;

wherein the conditions in the initial reaction stage provide a stable heterogeneous reaction mixture.

117. (Previously Presented) A process for the synthesis of polyol fatty-acid polyesters by reacting a polyol and a fatty-acid lower-alkyl ester under substantially solvent free conditions in the presence of a catalyst and an emulsifier, the process comprising:

an initial reaction stage which is carried out under such conditions that the reaction mixture in said initial stage is in steady-state, with continuous introduction of reactants comprising polyol and fatty-acid lower-alkyl ester, and continuous removal of products comprising reaction mixture having a degree of esterification of about 10% or more and volatile alcohol formed during the initial reaction stage, and

one or more subsequent reaction stages in which the reaction mixture from said initial stage is further reacted to said polyol fatty-acid polyesters;

wherein the conditions in the initial reaction stage aid in solubilizing the polyol.

118. (Previously Presented) A process for the synthesis of polyol fatty-acid polyesters by reacting a polyol and a fatty-acid lower-alkyl ester under substantially solvent free conditions in the presence of a catalyst and an emulsifier, the process comprising:

an initial reaction stage which is carried out under such conditions that the reaction mixture in said initial stage is in steady-state, with continuous introduction of reactants comprising polyol and fatty-acid lower-alkyl ester, and continuous removal of products comprising reaction mixture having a degree of esterification of about 10% or more and volatile alcohol formed during the initial reaction stage, and

one or more subsequent reaction stages in which the reaction mixture from said initial stage is further reacted to said polyol fatty-acid polyesters;

wherein the conditions in the initial reaction stage aid in solubilizing the polyol and provide a stable heterogeneous reaction mixture.